

10773,822

RECEIVED  
CENTRAL FAX CENTER

OCT 03 2006

Description of the Invention

Applicant's invention relates to a method for preparing membrane electrode assemblies (MEAs), and in particular to a method of manufacturing a proton-conducting cation-exchange electrolyte membrane for use in a membrane electrode assembly (MEA), in which atmospheric pressure plasma deposition is used to deposit catalysts such as platinum onto a polymer substrate, or a substrate including carbon cloth or carbon particles. The invention has two principal characteristics:

- 1) The noble metal catalyst is deposited on the membrane by discharge enhanced chemical vapor deposition (PECVD); and
- 2) The PECVD is carried out at atmospheric pressure, without adding noble gases to the PECVD carrier gas.

REMARKS

Claims 1, 3-9, and 11-16 are pending, and stand rejected.

Response to the Examiner's Response to Applicant's Arguments:

1. Applicant notes that the previous remarks of 6/16/2006 with respect to the Fornsel reference have been fully considered and are persuasive, and therefore the rejection has been withdrawn.
2. The Examiner is now citing the Fukuda reference (US 6,849,306) as a substitute for the previously cited Fornsel reference – primarily to show that nitrogen, in addition to the noble gases, can be used as the carrier gas in an atmospheric plasma deposition.

New cited reference: Fududa (US 6,849,306)

The Fududa reference describes an atmospheric plasma deposition process using a very high frequency voltage exceeding 100 kHz. (Col 2, lines 9-16) The high frequency voltage process tends to leads to a rapid increase in contamination to the electrode, so the inventors developed an apparatus in which the reactive gas is not directly in contact with

10773.822

the surface of either electrode (Claim 1). This process is the opposite of Applicant's process (paragraph [0022] and Figures 1A and 1B in which the reactants are passed directly between the electrodes. The Fududa reference describes the use of an inert gas such as "helium, neon, argon, krypton, xenon, or radon belonging to group XVIII in the periodic Table. In order to obtain the effects of the invention, helium or argon is preferably used." (Col. 12, lines 9-13) This statement is followed by the add-on "Nitrogen can be also used as the inert gas." (Col 12, line 13). In further listing components that can be additional parts of the carrier gas, mention is made of "nitrogen" (Col 12, lines 20-26). All of the Examples use either argon or helium as the carrier gas. The Examiner contends that the mention of nitrogen as a possible carrier gas, disregarding that it is in a secondary list, disregarding that it is not a preferred embodiment, disregarding that is not shown in any examples, and disregarding that it (but no noble gas) is included in gases that may be used in addition to the inert gases - would motivate one in the art to select nitrogen. Plus, the examples use only helium and argon - which clearly teaches away from Applicant's claims. Applicant contends that the overall teaching of the Fududa reference, teaching away from Applicant's "no noble gas in the carrier gas" and requiring that the reactive gas not be directly in contact with the electrodes (opposite Applicant's disclosure and drawings, would not motivate one in the art to arrive at Applicant's claims.

The Fududa reference fails to teach or suggest a proton-conducting cation-exchange electrolyte membrane for use in a membrane electrode assembly (MEA).

Even if the Fududa reference did not teach away from Applicant's claims, there is no motivation to combine the atmospheric process of Fududa with the vacuum or low pressure processes of Dearnley, Allen or Kirk-Othmer. While the Kirk-Othmer teaches a preferred low frequency process (page 5, line 8 of the first full paragraph) the Fududa process uses very high frequency process.

Summary of the cited references: Applicant's claim 1 requires:

A method of manufacturing a proton-conducting cation-exchange electrolyte membrane for use in a membrane electrode assembly (MEA), comprising the step of depositing at least one catalyst layer directly onto a substrate by passing reactants

10773,822

included in a carrier gas through an electrical discharge at atmospheric pressure, wherein said substrate is selected from the group consisting of a polymer membrane, a membrane formed of carbon cloth, and a membrane including carbon particles, and wherein the deposition is carried out without adding a noble gas to the carrier gas. (Discharge enhanced chemical vapor deposition - DECVD)

- a) The Dearnley reference describes a chemical vapor deposition of a catalyst for a fuel cell electrode in a vacuum system, and involves no carrier gas. It **does not teach or suggest atmospheric pressure, DECVD or a non-noble carrier gas.**
- b) The Allen reference describes a dual ion-beam assisted deposition to form electrodes in a vacuum. It **does not teach or suggest atmospheric pressure, or DECVD, or a non-noble carrier gas.**
- c) The Kirk - Othmer reference describes a plasma enhanced chemical vapor deposition (PECVD) process for making coatings (not catalysts) which is typically done in vacuum or very low pressure. It **does not teach or suggest DECVD, catalyst deposition or the formation of an MEA.** It also teaches away from an atmospheric process.
- d) The Fukuda reference describes an atmospheric very high frequency voltage DECVD process in which the reactive gas does not contact the electrode. Noble gases are listed, preferred and exemplified as the carrier gas, though nitrogen is also listed as a possible inert gas. It **teaches away from a carrier gas not containing a noble gas.**

The Examiner states that Applicant has remarked how each of the listed references fails to teach or suggest Applicant's claims, but has not addressed them in combination. Applicant has shown the deficits of each reference, and has shown how the added references fail to heal the deficits in the other references.

The Examiner then picks specific elements

- deposition of a catalyst for an electrode from Dearnley and Allen

10773,822

- “inert” vs. noble gas from the Allen background, and also since CVD and sputtering and other deposition methods are mentioned they must all be equivalent.
- and the use of an atmospheric discharged enhanced process from Fukuda to create Applicant’s claims.

According to MPEP 2143.01, the fact that references can be combined or modified is not sufficient to establish *prima facie* obviousness, unless the prior art also suggests the desirability of the combination. The Examiner has painfully taken many unrelated references from unrelated art areas, teaching a multitude of different methods for different purposes. Picking and choosing specific elements from various prior art references to create the claimed invention is not proper §103 analysis. Applicant contends that the obviousness rejection in this case is a classic example of hindsight, in effect using Applicant’s claims as a template on which selected bits of prior art teachings can be assembled. This is not a proper basis for rejection of claims. “One cannot use hindsight reconstruction to pick and choose among isolated disclosures in the prior art to deprecate the claimed invention.” *In re Fine*, 5 U.S.P.Q. 1596, 1600 (Fed. Cir. 1988).

Each of the cited references involving catalyst deposition to form an electrode is performed by a method (CVD or PECVD) than that claimed by Applicant, all at reduced pressure or vacuum. These processes, with their deficiencies described in Applicant’s background section of the Specification, is NOT the same as the DECVD process claimed by Applicant, and one in the art would not be motivated by disclosure of a CVD or PECVD process to try Applicant’s claimed DECVD process. These other processes not only fail to teach or suggest a DECVD process, but they teach away from a DECVD process by describing only other deposition methods. The lone reference to an atmospheric discharged enhanced process relates to a very high frequency voltage process for plasma treatment in which the reactive mixture does not directly contact the electrode. This is the opposite of Applicant’s described and illustrated apparatus in which the electrodes are directly in contact with the reactive mixture, and has nothing to do with, nor would it motivate one in the art to practice Applicant’s claimed process of a method of manufacturing a proton-conducting cation-exchange electrolyte membrane for

10773,822

use in a membrane electrode assembly (MEA), comprising the step of depositing at least one catalyst layer directly onto a substrate by passing reactants included in a carrier gas through an electrical discharge at atmospheric pressure, wherein said substrate is selected from the group consisting of a polymer membrane, a membrane formed of carbon cloth, and a membrane including carbon particles, and wherein the deposition is carried out without adding a noble gas to the carrier gas.

35 U.S.C. §103

Dearnley in view of Allen and further in view of Kirk-Othmer and Formsel

Claims 1, 3, 5-6 and 11-13 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Allen (US Patent Number 6,077,621) and further in view of Kirk-Othmer and Fukuda (US 6,849,306). The references fail to teach or suggest all of Applicant's claim limitations, as amended, thus no *prima facie* case of obviousness is presented. Specifically, the references fail to teach or suggest a method for manufacturing a cation-exchange membrane by DECVD, where the deposition is carried out at atmospheric pressure without adding a noble gas to the DECVD carrier gas, as set forth in Applicant's previous response.

Dearnley

The Dearnley reference discloses a vapor deposition method for depositing a catalyst on a fuel cell electrode under vacuum. The Dearnley reference fails to disclose Applicant's method using a carrier gas, and at atmospheric pressure. The Dearnley reference is silent on not adding a noble gas to the carrier gas – since no carrier gas is involved. The Dearnley reference not only fails to teach or suggest two critical limitations in Applicant's claims (carrier gas and an atmospheric pressure process), it teaches away from Applicant's claims by requiring a vacuum deposition and void of a carrier gas. One in the art would not be motivated to practice Applicant's atmospheric, carrier gas method from the vacuum process without a carrier gas disclosed in the Dearnley reference.

10773.822

Allen

The Allen reference describes a process for preparing an ion-conducting membrane by subjecting under vacuum the membrane to a low energy electron beam. For cleaning the membrane, then subjecting under vacuum the membrane to a high energy electron beam containing metal ions. As with the Dearnley reference above, the process of Allen is a vacuum process. One in the art would not be motivated to practice Applicant's atmospheric, carrier gas method from the vacuum process without a carrier gas as disclosed in the Allen reference.

Applicant believes the Examiner cites the Allen reference, not for the specific process taught by Allen which is not done at atmospheric pressure, but rather for the description in the Allen reference of the State of the Art. The Allen background describes methods of depositing heavy metal ions that include ultra-high vacuum (UHV), evaporation, chemical vapor deposition (CVD) and sputter deposition. The Examiner concludes that these methods are substitutes for each other, and thus CVD could be used to achieve a similar result to that of the Dearnley reference. However, while column 2, lines 58-65 describe a CVD process at atmospheric temperature, it also points out that the constituents of a vapor phase are often diluted with an inert gas. Applicant claims a discharged enhanced chemical vapor deposition which is a different process than a CVD process such as that described by Allen. Further, while the Allen reference points out that a CVD process uses an additional inert carrier gas, Applicant's claimed process is "without adding noble gasses to the DECVD carrier gas. One of skill in the art would not be motivated to practice the DECVD process of Applicant's claims from a reference to a CVD process, and most certainly would not be motivated to practice Applicant's claims of no added noble gas from a description of the use of an inert carrier gas. The Allen teaching of an inert carrier gas is exactly opposite and teaches away from Applicant's claims of no added noble gas.

The Examiner makes a point that the Allen reference – in describing background art, mentions a chemical vapor deposition (CVD). The mention of an inert carrier gas in column 2, line 61 is a passing reference to processes in the art without detail to actual gases used. All specific references known to Applicant of a CVD process involve a noble gas in addition to an inert gas such as N<sub>2</sub> or H<sub>2</sub>. While Applicant agrees

10/773,822

with the Examiner that all noble gases are inert, but not all inert gases are noble, the main points are that the passage cited in the Allen reference is not to the Allen invention, but rather to cited background art that is not described in detail. No fully described background art known by Applicant does not include a noble gas in the inert gas mixture.

More importantly, the Allen reference describes chemical vapor deposition (CVD), and its disadvantage of high temperature. It does not mention discharge enhanced chemical vapor deposition (DECVD) (as claimed by Applicant) which overcomes the temperature issue. CVD (Allen) and DECVD (Applicant) are different processes. Thus the Allen reference is incomplete in its background description by not better defining the composition of the "inert carrier gas", and describes the problematic CVD process rather than Applicant's DECVD process. Therefore the Allen reference does not teach or suggest Applicant's DECVD process – therefore does not alone present a *prima facie* case of obviousness, nor does it heal the other cited references which also fail to teach or suggest a DECVD process for the deposition of a catalyst layer.

#### Kirk-Othmer

The Kirk-Othmer reference is a secondary reference used to show plasma enhanced chemical vapor deposition. The Kirk-Othmer reference deals primarily with methods for depositing coatings such as Si, Al, Ti, B, W, and not with the deposition of a catalyst, as claimed by Applicant. Moreover, the Kirk-Othmer reference points out that plasma CVD is best at low gas pressure (page 5, first full paragraph). There is nothing in the Kirk-Othmer reference to motivate one to combine atmospheric pressure plasma deposition of a catalyst by DECVD without adding noble gases to the carrier gas. And the Kirk-Othmer reference also fails to heal the deficiencies of Dearnley and Allen teaching a low pressure or vacuum process rather than one at atmospheric temperature. Indeed the teaching of Kirk-Othmer of a preferred low pressure process teaches away from Applicant's claim of an atmospheric pressure process.

#### Fukuda

10/773,822

The Fukuda reference is described in detail above. It teaches away from Applicant's process and also teaches away from a carrier gas not containing a noble gas. It would not motivate one to practice Applicant's claims.

Dearnley in view of Allen and further in view of Kirk-Othmer and Fukuda and further in view of Schutze

Claim 4, stands rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Allen (US Patent Number 6,077621 and further in view of Kirk-Othmer and Fuduka, and further in view of Schutze. The Schutze The Schutze reference teaches a plasma jet using flowing helium. Applicant's amended claims cite a method without adding noble gases to the DECV carrier gas. The Schutze reference not only fails to teach or suggest Applicant's claim limitation of no added noble gas, but teaches away from Applicant's claims by requiring a noble gas. The Schutze reference fails to correct the deficiencies of the Dearnley reference, fails to teach or suggest Applicant's claim limitations, and teaches away from Applicant's claims.

Further in view of Yasumoto

Claim 7 stands rejected further in view of Yasumoto (US 2003/0096154). The Yasumoto is a secondary reference cited by the Examiner to teach the spraying of the catalyst onto the surface of the polymer electrode membrane. Applicant's do not claim a method in which a catalyst is sprayed onto a polymer electrode membrane, but rather a discharge enhanced chemical vapor deposition method. Thus the Yasumoto reference fails to teach Applicant's claims.

Further in view of Nanaumi

Claims 8-9 stand rejected under 35 U.S.C. 103(a) as being unpatentable further in view of Nanaumi (US 2004/0180250).

10773,822

The Nanaumi reference is cited to cite polymer electrolyte membrane structures. However the Nanaumi reference fails to teach or suggest Applicant's many claim limitations, and fails to correct the many deficiencies of the other references cited.

Further in view of Kamo

Claims 14 and 15 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Schutze in view of Fornsel (WO 01/32949, US 6,800,336), and further in view of Kamo (US 2003/0059659). The Kamo reference is a secondary reference cited to show the use of a platinum alloy in the anode side of an electrolyte membrane. While the Kamo reference discloses a platinum/ruthenium alloy for a fuel cell electrode, the platinum/ruthenium alloy is supported on a carbon powder, rather than directly on a membrane as claimed by Applicant. In Example 2, the platinum/ruthenium alloy is screen printed using a slurry. One in the art would not be motivated by this method alone – or in combination with the other cited reference to practice all of the limitations in Applicant's amended claims.

Further in view of Haug

Claim 16 stands rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Schutze in view of Fornsel (WO 01/32949, US 6,800,336), and further in view of Haug. The Haug reference is a secondary reference cited to show the deposition of multiple catalyst layers. The Haug reference demonstrates the use of a vacuum sputter deposition system for producing a PEM. The disclosure of a multiple layer of catalyst by methods teaching away from Applicant's claimed method fails to heal the defects of the cited art to present a *prima facie* case of obviousness.

Conclusion

The references cited, either alone or in combination, fail to teach or suggest all of Applicant's claim limitations, and therefore fail to present a *prima facie* case of obviousness over Applicant's amended claims. For the above reasons the present claims

10/773,822

1, 3-9, and 11-16 are believed by the Applicant to be novel and unobvious over the prior art, thus the claims herein should be allowable to the Applicant. Accordingly, reconsideration and allowance are requested.

Accordingly, reconsideration and allowance are requested.

Respectfully submitted,

  
\_\_\_\_\_  
Thomas F. Roland  
Attorney for the Applicants  
Reg. No. 42,110  
Tel. 215-419-7314

Date: October 3, 2006

Customer Number 31684